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469478

From: STIC-ILL
Sent: Friday, October 24, 2003 9:40 AM
To: Kozumplik, Joanne (ASRC)
Subject: FW: 10038600

-----Original Message-----
From: Mellerson, Kendra
Sent: Friday, October 24, 2003 9:37 AM
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From: Gakh, Yelena
Sent: Thursday, October 23, 2003 6:23 PM
To: STIC-EIC1700
Subject: 10038600

Dear Kendra,

please order the following:

2. TITLE: Development of scanning microwave microscope for high-throughput characterization of combinatorial dielectric thin film
AUTHOR(S): Okazaki, Noriaki; Ahmet, Parhat; Chikyow, Toyohiro; Odagawa, Hiroyuki; Cho, Yasuo; Fukumura, Tomoteru;
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Thanks,

Yelena

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Development of Scanning Micr wave Microscope for High-Throughput Characterization of Combinatorial Dielectric Thin Film

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ABSTRACT

A scanning microwave microscope (SμM) for high-throughput characterization of combinatorial dielectric materials has been developed using a lumped constant resonator probe. The probe consists of a microwave oscillator module equipped with a thin conducting needle and an outer conductor ring, which detects the dielectric constant of the sample just beneath the needle as a frequency shift of the resonator. The quantitative analysis of the dielectric constant for the bulk and the thin-film samples was carried out based on the measurement of gap-length dependence of the frequency shift. The analysis method was successfully applied to the characterization of composition-spread $Ba_xSr_{1-x}TiO_3$ thin film sample. The evaluation of far-field contribution to the frequency shift was found to be crucial for the accurate determination of dielectric constant especially in the characterization of combinatorial thin films.

INTRODUCTION

High throughput characterization of electric properties such as dielectric constant and loss tangent is highly required for the combinatorial design of dielectric materials. Especially, quantitative evaluation of these properties in the rf / microwave frequency regions has been increasing its importance in the field of communication and information technologies. For this purpose, the scanning microwave microscope (SμM) has been intensively studied in recent years.

The implementation of SμM designed so far is formally divided into two categories according to the style of resonator probe. One is the coaxial cavity resonator probe [1-7] and the other is the LC (inductance and capacitance) lumped-constant resonator probe [8-15]. The measurement principle of the (linear) dielectric constant is nearly the same for the two systems, in which the dielectric constant beneath the probe needle is detected as a shift of the resonance frequency. Advantage of the former resonator is its stability and high quality factor. Quantitative measurements of dielectric constant, loss tangent and surface conductivity have been made and its application to the characterization of combinatorial library samples has been reported [1-7]. The latter system has so far been used most predominantly to measure the nonlinear dielectric constant with applying additional low frequency AC voltage to the sample. Domain structure of dielectric materials has been successfully observed by this microscope with 1 nm resolution [11,12,14]. It has also been shown that the LC resonator probe is readily mounted on the atomic

force microscope (AFM) to obtain the sample morphology and dielectric constant simultaneously [14]. The most advantageous point in the latter system is the extremely small probe size, which makes the system simpler, and practical operations such as coarse approaching and scanning much easier. We have recently developed a SμM system using the LC resonator probe, which has been shown to possesses enough ability for the high-throughput characterization of the combinatorial dielectric samples [15].

In this paper, we report on the quantitative analysis method based on the measurement of gap-length dependence of the frequency shift. The validity of the method has been checked using several standard bulk and thin-film samples. It will be shown that the analysis can be applied straightforward to the characterization of combinatorial samples if the far-field contribution to the frequency shift is adequately taken into account. The analysis procedure is described in detail using the measured data of composition-spread $Ba_xSr_{1-x}TiO_3$ thin film.

EQUIPMENT

The LC resonator probe was fabricated using a commercially available microwave oscillator module with a free carrier frequency of ~1.2 GHz (Murata, Japan). The probe needle was obtained by electrochemically polishing a tungsten wire and was attached to the oscillator module. Typical size of the needle tip was *ca.* 20 μmϕ. Full dimension of the resonator probe was as small as $7 \times 5 \times 1$ mm^3. The probe module was carefully shielded to minimize undesirable background frequency shift due to a far field contribution (discussed below). The probe was installed on a vertical linear-motion guide equipped with a balance arm (Chuo Seiki, Japan), which guarantees constant contact pressure during the scanning operation without loosing the accuracy of lateral positioning of the needle. The balanced probe unit was mounted on a mechanical Z stage (Sigma, Japan) for automatic needle approach operation. The lateral scanning of the sample relative to the probe was carried out by a mechanical XY stage (Sigma, Japan) mounting the sample. The scanning area is as large as 20×20 mm^2, which is suitable for the characterization of combinatorial samples, with the minimum step size of 1 μm. The frequency measurement was performed by a spectrum analyzer (Advantest, Japan). Stage motion and data acquisition were totally computer-controlled through a GPIB interface (Interface, Japan).

ANALYSIS METHOD

If a sample is set sufficiently far away from the LC lumped-constant resonator probe, its free carrier frequency f_0 is given by

$$f_0 = \frac{1}{2\pi\sqrt{LC_0}}, \tag{1}$$

where L and C_0 represent the inductance and the stray capacitance between the needle and the electric circuit of the probe, respectively. When the tip approaches the sample surface, the carrier frequency shifts by Δf_r due to the small capacitance between the needle and the outer conductor ring (C_s) connected in parallel to the original LC circuit.

$$f_0 + \Delta f_r = \frac{1}{2\pi\sqrt{L(C_0 + C_s)}} \tag{2}$$

Since C_s is much smaller than C_0, the normalized frequency shift $-\Delta f_r/f_0$ is approximated as

$$-\frac{\Delta f_r}{f_0} = \frac{1}{2}\frac{C_s}{C_0}. \tag{3}$$

For the bulk sample in the contact mode, the capacitance C_s can be calculated explicitly as

$$C_s = -4\pi\varepsilon_0 R_0\left(\frac{\ln(1-b)}{b}+1\right) \text{ with } b = \frac{\varepsilon_r - 1}{\varepsilon_r + 1}, \tag{4}$$

where R_0 is the effective tip radius of the needle, and ε_r is the relative dielectric constant of the sample, respectively. Substituting eq. (4) into eq. (3), we get

$$-\frac{\Delta f_r}{f_0} = A f(b), \tag{5}$$

where

$$A = 2\pi\varepsilon_0 R_0 / C_0 \tag{6}$$

and

$$f(b) = -\left(\frac{\ln(1-b)}{b}+1\right). \tag{7}$$

In eq. (5), only the unknown parameter is A, indicating that the dielectric constant of any sample can be determined if the frequency shift is calibrated against one standard sample with known dielectric constant [13].

However, in the actual measurement (especially for the sample with small dielectric constant), we found that "non-tip-end" capacitance significantly contributes to the measured frequency shift (far field contribution), which was unavoidable despite the careful shielding of the resonator-probe unit. For the accurate evaluation of dielectric constant, eq. (5) should be augmented by the non-tip-end contribution F.

$$-\frac{\Delta f_r}{f_0} = A f(b) + F, \tag{8}$$

In this situation, the relative measurement using the standard sample is not applicable since F may differ significantly from one sample to another, and it can also vary with the measurement point even within a single sample. The best and practical way to overcome the difficulty is to measure the frequency shift as a function of the air gap length (g) in the vicinity of the contact point, where near field contribution becomes large while F remains virtually constant. In the presence of air gap, the function $f(b)$ in eq. (8) should be replaced by $f(b, g/R_0)$, which has been solved rigorously using the image charge method [6]. The unknown parameters A, R_0, F and ε_r can be obtained simultaneously by the curve fitting. However, it is better to determine at least R_0 from the standard sample measurement in advance, and to fix it throughout the unknown sample measurements in order to increase fitting accuracy.

The same analysis procedure can be applied to the determination of thin-film dielectric constant, although the accuracy may be somewhat lowered due to the large contribution of the substrate material. In this case, the function f in eq. (8) should be replaced by $f(\varepsilon_s, \varepsilon_f, t/R_0, g/R_0)$, where ε_s and ε_f represent the dielectric constants of the substrate and the thin film, respectively, t/R_0 denotes the normalized film thickness and g/R_0 is the normalized gap length. Although the explicit form of the fitting function cannot be obtained for this configuration, approximate formulation has so far been made either by the image-charge method [6] or by the numerical simulation based on the finite element method [7]. The parameters ε_s, t and R_0 should be

determined in advance from the separate measurement. Remaining parameters to be determined by the fitting calculation are A, F and ε_f.

RESULTS AND DISCUSSION

Figure 1 shows the gap-length dependence of the frequency shift measured for several samples. First we measured $LaAlO_3$ ($\varepsilon_r = 24$) as a standard sample to determine the effective tip radius R_0. As can be seen from the data (Fig. 1 (a)), significant frequency shift occurs only in the vicinity of contact point of the order of ~1 μm. Therefore, enough resolution is required for the vertical motion in order to achieve high accuracy in the fitting calculation. In the present experiment, we set the Z-stage step size at 0.2 μm. For the fitting calculation, we used the fitting functions proposed by Lee *et al.* [7], and Levenberg-Marquardt method was employed as a fitting algorithm. The measured data was well fitted by the fitting function for bulk case [7]. The obtained value of R_0 was consistent with the optical microscopic observation. Figure 1 (b) and (c) show the data for MgO and $LiNbO_3$ bulk samples, respectively. From the fitting calculation (with R_0 being fixed), we obtained dielectric constants close to the literature values for both cases. Figure 1 (d) shows the result for $SrTiO_3$ thin film deposited on $LaAlO_3$ substrate. We used the fitting function for the thin film on $LaAlO_3$ (Case-C in ref. 7). The obtained dielectric constant of

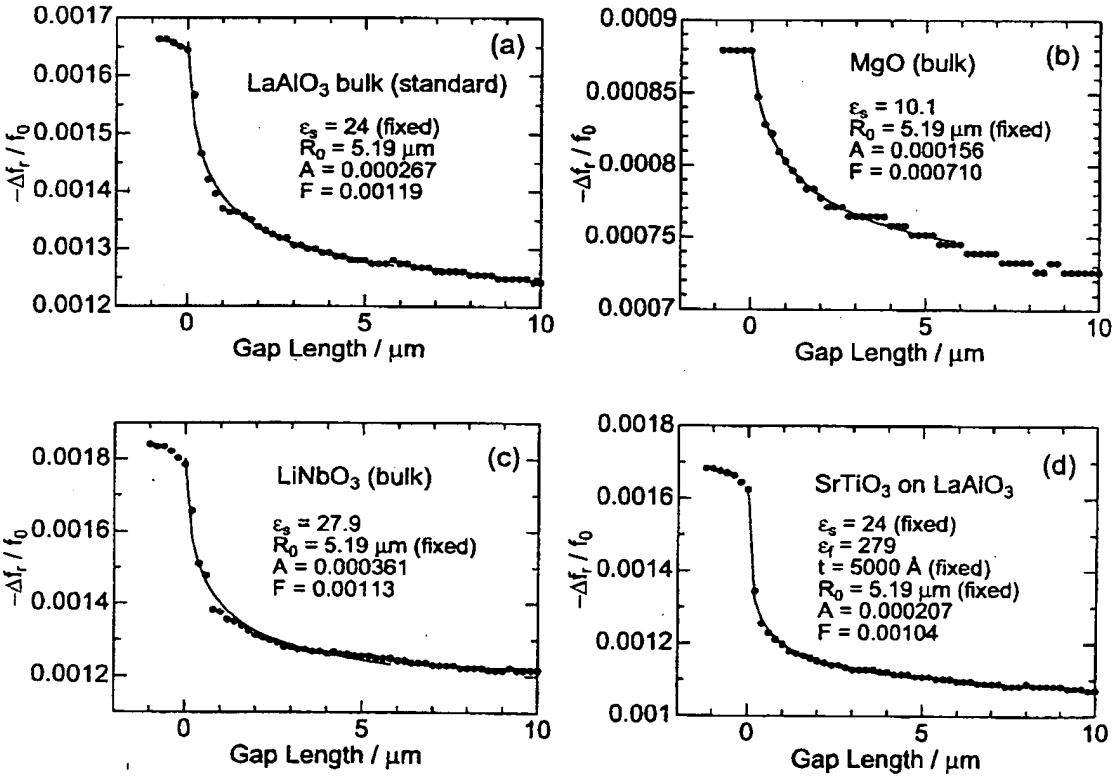

Figure 1. Gap-length dependence of the relative frequency shift measured by SμM. Solid line shows the fitting results.

the $SrTiO_3$ thin film is close to the literature value (292) estimated by the cavity- type microwave microscope [6]. All the results described above support the general applicability of the analysis method in combination with our SμM system.

We have further applied the present analysis method to the mapping of dielectric constant for a combinatorial thin-film sample. Figure 2 (a) shows the line-scan data obtained for the composition-spread $Ba_xSr_{1-x}TiO_3$ thin film deposited on $LaAlO_3$ substrate. As discussed in the previous section, the far-field contribution can vary depending on the measurement point even in a single sample. Assuming that F is a smooth function of the lateral position, we performed the gap-length dependence measurement for the selected operation points with the interval of 1 mm. The far-field component F obtained by the fitting calculation is shown in Fig. 2 (b), which shows decreasing tendency with lateral position. The parameter A simultaneously obtained by the fitting procedure did not show the significant dependence on the lateral position. Note that R_0 was determined in advance from the separate gap-length dependence measurement for the marginal substrate area of the same sample. The profile of F was, after fitted by the smooth function, subtracted from the original frequency shift data. Finally, the remaining part $-\Delta f_r / f_0 - F$ was converted to the thin-film dielectric constant through the contact-mode fitting function. The profile of thin-film dielectric constant (ε_f) is given in Fig. 2 (c), which shows similar tendency with the data previously reported for the $Ba_xSr_{1-x}TiO_3$ thin film on Nb-doped $SrTiO_3$ substrate [15].

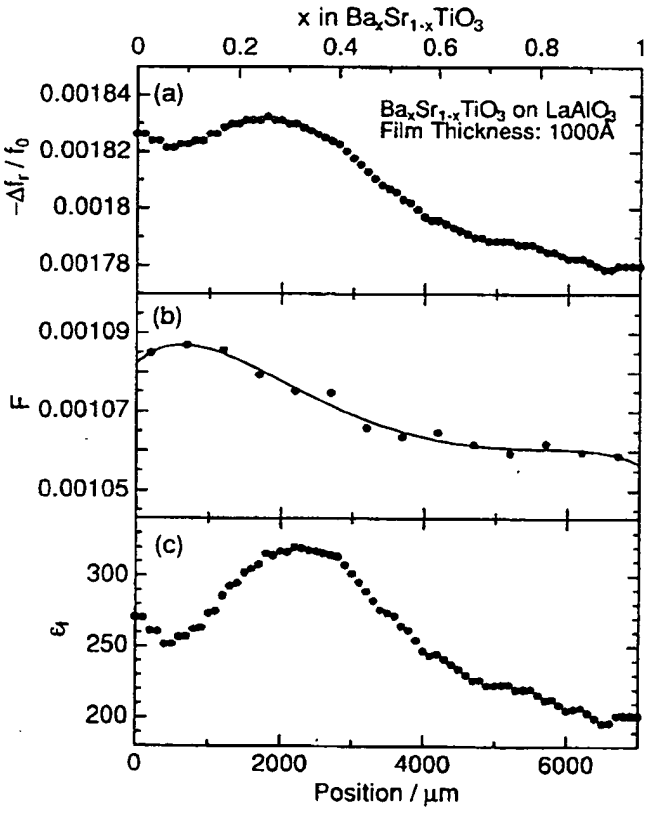

Figure 2. Line profiles of (a) frequency shift raw data, (b) far-field component and (c) dielectric constant measured for the composition-spread $Ba_xSr_{1-x}TiO_3$ thin film.

CONCLUSIONS

In summary, we developed the scanning microwave microscope using a LC lumped-constant resonator probe. The quantitative analysis of dielectric constant was carried out based on the measurement of gap-length dependence for bulk and thin-film cases, through which validity of the analysis method was confirmed. The method was applied to map the distribution of dielectric constant in the composition-spread $Ba_xSr_{1-x}TiO_3$ thin film. The evaluation of the far-field component and its dependence on the measurement position was shown to be crucial for the accurate mapping of dielectric constant in the combinatorial thin film samples.

REFERENCES

1. Y. Cho, A. Kirihara and T. Saeki, *Rev. Sci. Instrum.* **67**, 2297 (1996).
2. T. Wei, X.-D. Xiang, W. G. Wallace-Freedman and P. G. Schultz, *Appl. Phys. Lett.* **68**, 3506 (1996).
3. Y. Lu, T. Wei, F. Duewer, Y. Lu, N. Ming, P. G. Schultz and X.-D. Xiang, *Science* **276**, 2004 (1997).
4. I. Takeuchi, T. Wei, F. Duewer, Y. K. Yoo, X.-D. Xiang, V. Talyansky, S. P. Pai, G. C. Chen and T. Venkatesan, *Appl. Phys. Lett.* **71**, 2006 (1997).
5. H. Chang, C. Gao, I. Takeuchi, Y. Yoo, J. Wang, P. G. Schultz, X.-D. Xiang, R. P. Sharma, M. Downes and T. Venkatesan, *Appl. Phys. Lett.* **72**, 2185 (1998).
6. C. Gao and X.-D. Xiang, *Rev. Sci. Instrum.* **69**, 3846 (1998).
7. J. H. Lee, S. Hyun and K. Char, *Rev. Sci. Instrum.* **72**, 1425 (2001).
8. Y. Cho, S. Atsumi and K. Nakamura, *Jpn. J. Appl. Phys.* **36**, 3152 (1997).
9. Y. Cho, K. Matsuura and J. Kushibashi, *Jpn. J. Appl. Phys.* **37**, 3132 (1998).
10. Y. Cho, K. Matsuura, S. Kazuta, H. Odagawa and K. Yamanouchi, *Jpn. J. Appl. Phys.* **38**, 3279 (1999).
11. Y. Cho, S. Kazuta and K. Matsuura, *Jpn. J. Appl. Phys.* **38**, 5689 (1999).
12. Y. Cho, S. Kazuta and K. Matsuura, *Appl. Phys. Lett.* **75**, 2833 (1999).
13. Y. Cho, S. Kazuta, K. Ohara and H. Odagawa, *Jpn. J. Appl. Phys.* **39**, 3086 (2000).
14. H. Odagawa, Y. Cho, H. Funakubo and K. Nagashima, *Jpn. J. Appl. Phys.* **39**, 3808 (2000).
15. N. Okazaki, H. Odagawa, Y. Cho, T. Nagamura, D. Komiyama, T. Koida, H. Minami, P. Ahmet, T. Fukumura, Y. Matsumoto, M. Kawasaki, T. Chikyow, H. Koinuma and T. Hasegawa, *Appl. Surf. Sci.*, in press.